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An investigation on the variations occurring during Ni₃Al powder formation by mechanical alloying technique

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ABSTRACT

The Ni₃Al intermetallic compound with unique properties is widely used especially for high temperature applications such as gas turbines. There are different ways for producing the compound. Among them, high-energy milling technique using a planetary ball mill has been employed for producing nanocrystalline Ni₃Al powders. In this research Ni₃Al intermetallic was prepared by mixing pure elemental Al and Ni powders. A ball-to-powder weight ratio of 20:1 and rotation speed of 550 rpm in argon atmosphere were considered as the main processing parameters. The milling time ranged from 1 to 55 h. Changes in the phase and microstructure as a function of milling time were investigated using X-ray diffraction analysis and scanning electron microscopy. The results revealed that the formation of nanocrystalline Ni₃Al with minimum crystallite size of 5 nm is obtained after 15 h milling. However, further mechanical alloying resulted in increasing grain size. Furthermore, the process efficiency decreased to less than 50% and the lattice strain increased after 55 h mechanical alloying.

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1. Introduction

Five intermetallic compounds such as: Al₃Ni, Al₃Ni₂, Al₃Ni₅, NiAl and Ni₃Al can be found in Ni–Al binary phase diagram. Ni₃Al with significant high strength, good oxidation and corrosion resistance at elevated temperature has been widely used in different applications. However, because of the extreme brittleness of Ni₃Al at room temperature there are many limitations in its application as an engineering material [1]. Fabrication of components from Ni₃Al through the conventionally casting method is difficult and expensive whereas powder metallurgy (PM) is a common method for fabricating high melting materials with enhanced mechanical properties.

A wide variety of methods including mechanical alloying (MA) are introduced for synthesis of nanostructured intermetallic compounds with ductile behavior. MA is a dry powder processing technique and has been used to synthesize both equilibrium and metastable phases of commercially useful and scientifically desirable materials. The technique was developed by Benjamin [2] to produce an alloy combining oxide dispersion strengthening with γ' precipitation hardening in a nickel-based superalloy designed for gas turbine applications. MA method is currently used in producing different materials such as intermetallic compounds.

The process of MA consists of several stages including loading powders and grinding medium (generally hardened steel or tungsten carbide balls) in a stainless steel container sealed under a protective argon atmosphere, in order to avoid/minimize oxidation and nitriding during milling, and milling for the predetermined period of time. Approximately, 1–2 wt.% of a process control agent (PCA) (usually stearic acid) is normally added to prevent excessive cold welding of the powder particles, especially when powders of ductile metals are milled [3].

Although some alloys and intermetallics synthesized by MA such as: Al–Cu [4], Al–Ni [5], Mn–Ni [6], Mg–Ni [7], Al–Ti [8], Fe–Al, Co–Al and Mn–Al [9] and Fe–Ni [10] have been studied earlier, there is less experimental results on mechanical alloying of Ni₃Al. The present work attempts to reveal the phase and microstructural changes occurring during milling of Ni and Al powders mixed in a nominal composition appropriate for Ni₃Al intermetallic.

2. Experimental procedures

2.1. Materials

Elemental Al powder (Merck-1056–99%, <160 μ m) and Ni powder (Merck-112277–99.5%, <10 μ m) were used as raw materials. The morphologies of both powders were examined using electron and optical microscopy.

2.2. Mechanical alloying process

Mechanical alloying of the elemental Al and Ni powders (with 1:3 atomic ratio of Al to Ni) was conducted using a planetary apparatus (model FP2) to produce Ni₃Al powder particles. The powders together with hardened steel balls with

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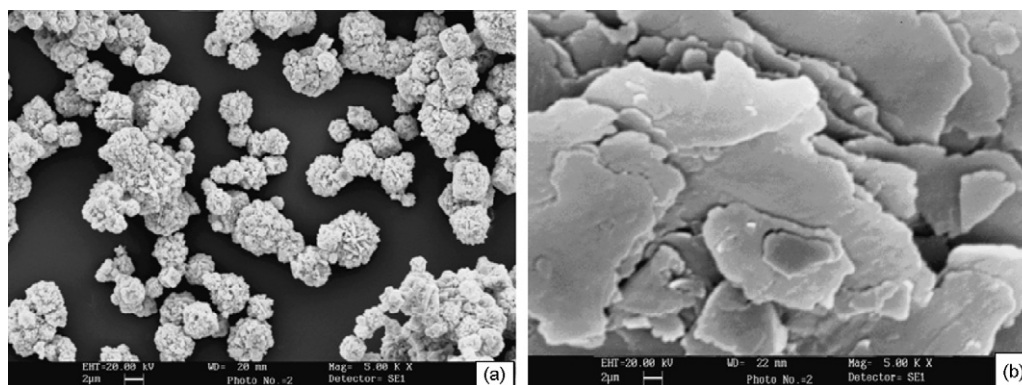


Fig. 1. Morphology of as-received (a) aluminum and (b) nickel powder.

20 mm in diameter were sealed in a stainless steel vial. The process was carried out at room temperature. The ball-to-powder weight ratio and rotational speed were 20:1 and 550 rpm, respectively. In order to avoid oxidation, the entire process was performed under argon atmosphere. After mechanical alloying for various time intervals, morphological and structural characteristics of the powders were analyzed.

2.3. Powder characterization

In order to determine the type and structure of Ni–Al compound formed during milling and to measure the crystallite size and lattice strain of particles the powders were characterized by Bruker; Advanced-D8 X-ray diffractometer. Moreover, morphological changes were examined using a scanning electron microscope model LEO 440i. The size distribution of powder particles was also evaluated by Clemex image analyzer.

The crystallite size and lattice strain were estimated through measuring the broadening of the X-ray peaks (Williamson–Hall integral breadth method (WH) [11]).

3. Results and discussion

Microstructural study of raw materials (Fig. 1) illustrates that the as-received Al powder is flake-like, whereas Ni powder is polygonal in shape. The powders react with each other during ball milling to make a Ni–Al compound. Ni and Al atoms tend to occupy certain sublattice sites of $L1_2$ and B_2 superlattice structures. Ni–Al compounds are one of the ordered intermetallic alloying systems. Diffusion of Al atoms into the Ni lattice during mechanical alloying process results in the formation of Ni(Al) solid solution and accordingly, Ni_3Al compound as a final product.

Fig. 2 shows X-ray diffraction patterns of powders obtained after different milling times from 1 to 55 h. At the early stages of milling, diffraction peaks are sharp.

During mechanical alloying, the sharp peaks of as-received powder are broadened progressively with increasing processing time due to the accumulated internal strain and refinement of grains. Meanwhile, the intensity of peaks decreases with mechanical alloying time as shown in Fig. 3.

As Fig. 3 indicates particles are attained at the first stage of MA. The decreasing rate of Al peak is higher than that of Ni, so that after milling for 5 h the intensity of Al peak begins to disappear. In comparison with Ni, since Al powder is very soft and can deform rapidly during alloying, broadening and disappearing of its peaks occur during the first hours of milling.

When milling continues, the impacts of balls on powder particles result in the diffusion of Al with atomic radius of 1.43 Å into the Ni lattice with the atomic radius of 1.24 Å. The difference between atomic radii causes expansion in the Ni lattice, as well as the displacement of its peaks to lower angles. Variation of lattice parameter is a sign indicating the formation of Ni_3Al . After 10 h of milling, Ni_3Al peak can be distinguished but its exact observation occurs at 15 h. da Silva and da Lima [12] reported the same

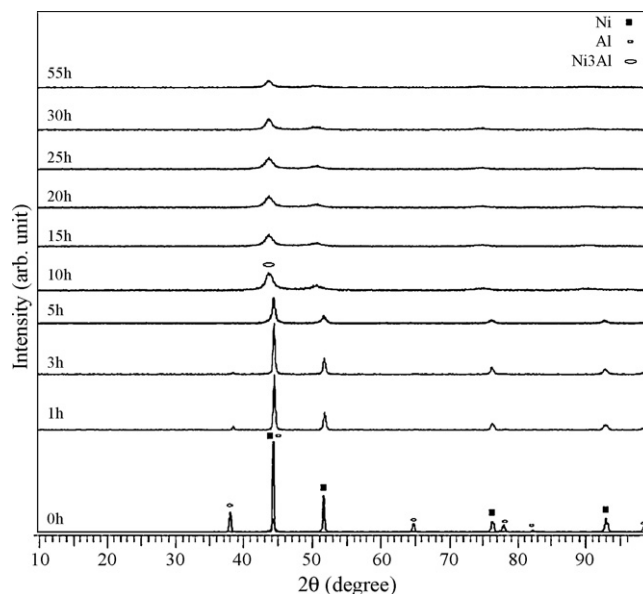


Fig. 2. X-ray diffraction patterns of the powders milled for different times.

results for Al–Cu powder after being mechanically alloyed at different times.

Fig. 4 shows SEM image of the powder at the milling time of 15 h indicating its homogeneity. The point analysis at different and random sites demonstrates that the atomic ratio of Ni to Al is 75:25. Back scattered image of powder confirms the results due to the fact that pure Al and Ni are not detected.

Variation of lattice parameter of Ni and Al during the milling process is indicated in Fig. 5. The lattice parameter was calculated from the XRD peaks using the Bragg's law, considering the fact that

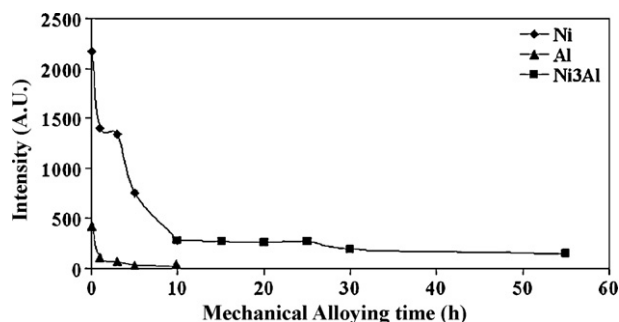


Fig. 3. Intensity of peaks versus milling time.

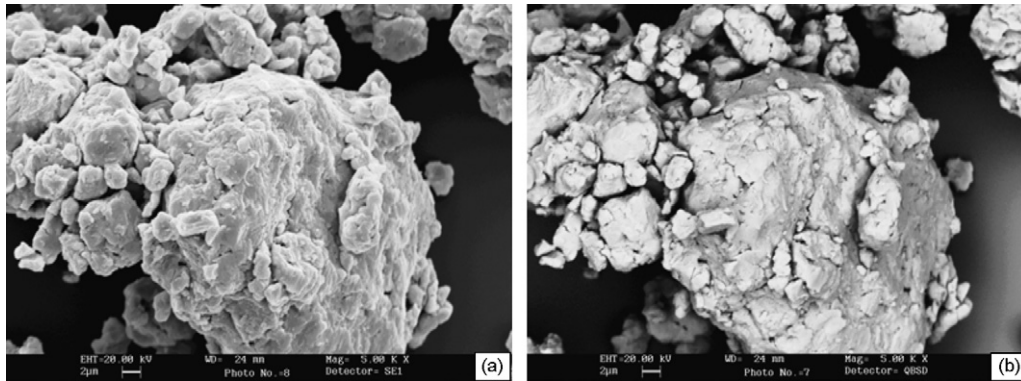


Fig. 4. SEM image of Ni and Al powder at the milling time of 15 h.

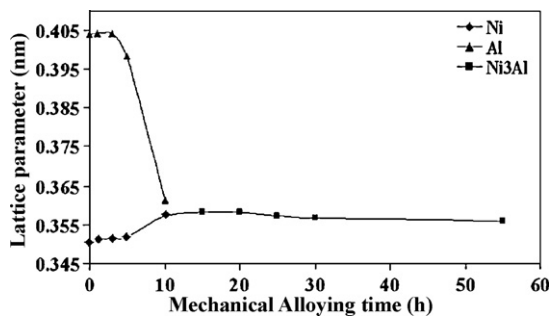


Fig. 5. Variation of lattice parameter of Ni and Al with milling time.

the interplanar distance of (*hkl*) planes (*d*) in fcc metals is related to the lattice parameter (*a*) by the following equation [13]:

$$d = \frac{a}{(h^2 + k^2 + l^2)^{0.5}} \quad (1)$$

Ni and Al powders are only mechanically bonded at first. Diffusive layers are extremely fine so that in some cases are not observed. This is widely accepted that the formation of the intermetallic compound using MA technique is carried out through a diffusion process in which atomic displacement occurs by vacancy mechanism [4]. This requires unoccupied nearest neighboring sites of an atom between Ni and Al elements. In addition, the atoms must have sufficient thermal energy to overcome the activation energy needed for migrating to the unoccupied sites. Longer milling time causes formation of defects in the lattice which in turn decrease the diffusion space.

Fig. 6 shows that the size of the 55-h MAed powder is larger than the powder milled for 15 h. During initial stages of milling,

sequences of welding and fracturing are perceived. The balance between them is achieved at 15 h. After 20 h of MA, welding dominates fracturing. Cold welding of powder particles occurs during long milling times causing an enlargement in the average particle size. Image analysis indicated that for the powder milled for 15 h the mean diameter of powders is 6.7 and the average aspect ratio is 1.96 while, for the powder milled for 55 h the magnitudes are 10.9 μm and 2.61, respectively. Intermetallic compounds are naturally brittle but the nanostructure obtained from mechanical alloying can cause more ductility in the material. Such evidence has been reported by Kumaran et al. [14] during high-energy milling of TiAl powders.

Impurity formation during long-time milling is one of the problems of mechanical milling. The impurities come from iron element which is a result of balls impacts. Fig. 7 shows entire EDS analysis of 15 and 55 h MAed powders. It is observed that the impurity has a considerable value for 55 h MAed powder. SEM study of the microstructures confirms the presence of fine Fe particles on the milled powder.

Fig. 8 exhibits variation of Ni and Ni₃Al powder crystallite size with different milling times. The value of crystallite size was estimated by measuring the broadening of the X-ray peaks. The peaks are related to Ni structure at the beginning (up to 10 h milling) and Ni structure transformed to Ni₃Al at the next stages (from 10 to 55 h milling). The powder attained nanocrystalline size (around 5 nm) at the early stages of milling (at 15 h of milling). This phenomenon is due to severe plastic deformation of particles which, in turn, is due to the multiplication of intergranular dislocations. By sliding and rearrangement of dislocations, a cellular structure with low angle boundaries is formed. This structure transformed to nano-sized grains during MA process.

Minimum grain size which is determined by balance between rate of dislocation generation, recovery and recrystallization is

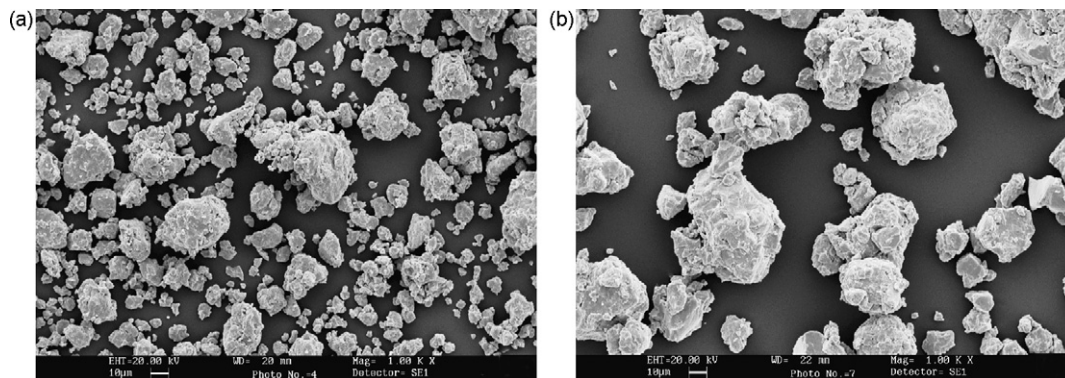


Fig. 6. SEM images of: (a) 15 h and (b) 55 h MAed powder.

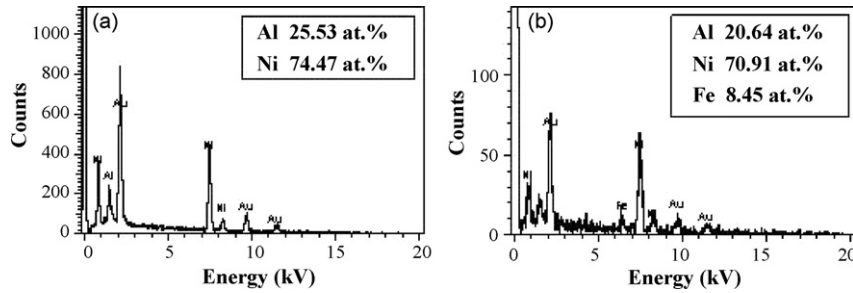


Fig. 7. EDX analysis of: (a) 15 h and (b) 55 h MAed powder.

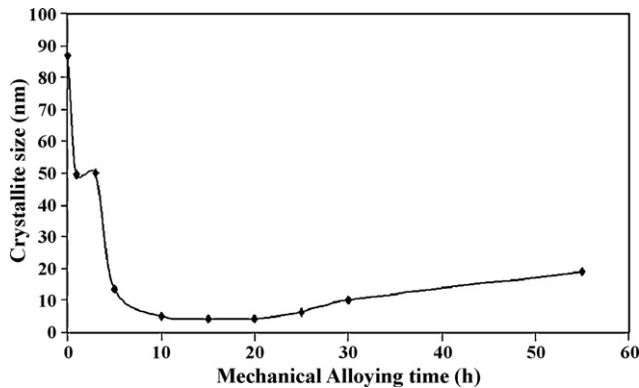


Fig. 8. Variation of Ni and Ni₃Al powder crystallite size with milling time.

achieved through this process. Refinement of grains acts as a limiting factor for continuation of the process.

During MAing of nanocrystalline powder grain boundary sliding or migration occurs. As a result, the nanocrystalline structure causes the ductility of Ni₃Al compound and increases cold welding which might be the reason for coarsening of intermetallic compound at higher milling times.

With increasing milling time, broadening of Ni₃Al peaks happens, however, there is no sign of amorphous structure even at 55 h of milling. Kumaran et al. [14] assert that this phenomenon could be mainly due to the fact that the energy supplied during milling is not sufficient to lower the free energy of the amorphous phase in Ni₃Al compound powder.

Continuous impact of balls at longer milling times results in the sticking of powder to balls and consequently, the process efficiency decreases. The observed decrease in the process effi-

ciency also implies the domination of welding on fracturing. Fig. 9 shows the process efficiency of the powders produced after different mechanical alloying times. The efficiency of process decreases with increasing mechanical alloying time, to less than 50% after 55 h mechanical alloying. It should be mentioned that the process efficiency at each milling time is defined as the ratio of acquired powder mass to the initial powder mass. Longer mechanical alloying may lead to a slight increase in grain size as a result of joining the nano-scale sub grains. The results designated that heat produced during mechanical alloying for longer than approximately 20 h, can promote grain growth and particle coarsening. Heating of powders causes further cold welding between particles and decreases the process efficiency.

The variation of lattice strain of the ball-milled powder with milling time is shown in Fig. 10. One peak is observed at 20 h of milling. It is reasonable to correlate the grain size produced during MA process with the lattice strain accumulated in the material. The figure shows that up to 20 h of MA the lattice strain increases while the grain size decreases.

The results obtained for the lattice strain are in good agreement with the grain size and the effect of ball impacts. Generally, the lattice strain increases with rising MA time. According to the similar grain size produced at 15 and 20 h milling, it is expected that the equal amounts of lattice strains are obtained in the time interval. However, an increase in lattice strain at 20 h MAing can be due to the further impact between balls and powder particles. Continuous impacts of balls and particles at longer milling times result in an increase in the lattice strain except for the milling time of 25 h. The heat created during Ni₃Al formation and impacts of balls to the powder particles can lead to such a decrease in the lattice strain. Variation of the lattice strain has been studied in several researches on mechanically alloyed intermetallics [15,16].

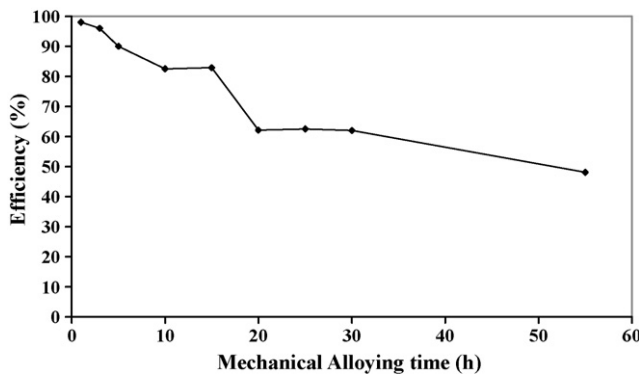


Fig. 9. Process efficiency of produced powder after different mechanical alloying time.

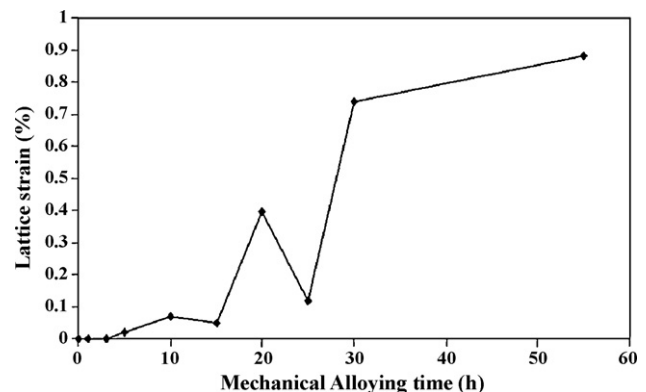


Fig. 10. Lattice strain of the ball-milled Al powders at different milling time.

4. Conclusions

The Ni₃Al intermetallic compound was produced using a planetary ball mill with the rotational speed of 550 rpm. Study of different characteristics of the compound at different milling times led to the following results:

- 1- The peak broadening analysis indicated that the crystallite size decreases rapidly at the beginning of milling (up to 15 h) but it slightly increases with further increasing milling time.
- 2- It was shown that the longer mechanical alloying time such as 55 h, causes to Fe contamination; while at the lower milling time (e.g. 15 h) impurities are not produced.
- 3- The process efficiency decreases to less than 50% after 55 h mechanical alloying.
- 4- It was indicated that the rate of increasing lattice strain at the beginning of MA up to 10 h is low and at 15 h milling the decrease in lattice strain occurs. Although, longer milling time causes higher values of lattice strain.

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